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<b>(21) International Application Number:</b> PCT/GB96/01033 <b>(22) International Filing Date:</b> 30 April 1996 (30.04.96)  <b>(30) Priority Data:</b> 281/95 4 May 1995 (04.05.95) YU 95/10534 12 December 1995 (12.12.95) ZA  <b>(71) Applicant (for MW only):</b> BOWMAN, Paul, Alan [GB/GB]; Braddan, Riding Lane, Hildenborough, Kent TN11 9LR (GB).  <b>(71)(72) Applicant and Inventor:</b> MARKOVIC, Miodrag [YU/YU]; RGI 32303 Brdani (YU).  <b>(74) Agent:</b> BOWMAN, Paul, Alan; Lloyd Wise Tregear & Co., Commonwealth House, 1-19 New Oxford Street, London WC1A 1LW (GB).		<b>(81) Designated States:</b> AL, AM, AT, AU, AZ, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, IS, JP, KE, KG, KP, KR, KZ, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TR, TT, UA, UG, US, UZ, VN, ARIPO patent (KE, LS, MW, SD, SZ, UG), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
<b>(54) Title:</b> PROCESS FOR PRODUCING INSULATING MATERIALS AND PRODUCTS THEREOF  <b>(57) Abstract</b>  A process for producing a thermal insulating calcium hydrosilicate compound comprises the steps of: providing a source of calcium ions; providing a silica compound; providing a stabilizing reagent; providing a fibrous compound; forming an aqueous mixture of the source of calcium ions the silica compound, the stabilizing reagent and the fibrous component; and hydrothermally treating the aqueous mixture at a pressure above 12,5 bars but not over 20 bars in the presence of saturated steam at a temperature from about 190 to 212 °C to produce a calcium hydrosilicate compound.		

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PROCESS FOR PRODUCING INSULATING  
MATERIALS AND PRODUCTS THEREOF

This invention relates to a process for producing thermal  
5 insulating materials which are resistant to high  
temperatures. More particularly it relates to a process  
for producing thermal insulating materials in the form of  
calcium hydrosilicate compounds. The invention also  
relates to products of the process.

10 It is well known that  $\text{CaO}$  and  $\text{SiO}_2$  react in the presence  
of water and at elevated temperatures to produce calcium  
hydrosilicates. The abbreviation C-S-H is hereinafter  
used for calcium hydrosilicate, where  $\text{C} = \text{CaO}$ ,  $\text{S} = \text{SiO}_2$ ,  
15 and  $\text{H} = \text{H}_2\text{O}$ .

Depending upon the chemical composition and particle size  
of the raw materials, their molar ratio, the amount of  
water present and the conditions of hydrothermal  
20 processing (pressure, temperature and time), a range of  
various crystal forms can be obtained of which  
tobermorite and xonotlite are the most significant ones.  
It is also known that C-S-H synthesis can be carried out  
by various processes.

25 The properties of the products obtained using these  
processes depend mostly upon the crystal form of the so  
produced hydrosilicate, the shape and size of crystals  
and the distribution and size of pores. To improve a  
30 hydrosilicate's mechanical properties, inorganic fibres  
(for example asbestos, glass or mineral) and/or organic  
fibres (for example cellulose) are frequently added.

USA Patent No 3,988,419 describes a process of C-S-H  
35 synthesis from an aqueous solution of very fine amorphous  
 $\text{SiO}_2$  (from waste dust out of a furnace for the production  
of Si, FeSi or SiC) and lime. The solution is heated for

some time at a temperature of 100°C under normal pressure while stirring to form a gel. The gel is then transferred into a rotary press where it is mashed slowly under pressure of saturated water steam of about 8kP/cm<sup>2</sup> until  
5 the gel is transformed into a viscous-elastic material which is poured into moulds. The moulds are then introduced into an autoclave and exposed to hydrothermal processing by means of saturated water steam under a pressure of about 20kP/cm<sup>2</sup> to form crystals. The material  
10 is then treated with overheated steam in order to allow the crystals to grow and also to dry the said crystals.

French Patent 2,505,814 describes a process which comprises the mixing of slaked lime, ground and ultra  
15 fine SiO<sub>2</sub> and synthetic fibres (allumosilicates, carbon) in an aqueous suspension. Moulds are then filled with the mixture which is then treated in an autoclave by means of saturated water steam under a pressure of about 12 bars and then dried for several days at the temperature of  
20 330°C.

USA Patents No. 3,895,096 and No. 4,467,041 also describe processes based on the same principle but using some other starting raw materials and fibres and with  
25 different CaO:SiO<sub>2</sub> mole ratios to obtain products with different crystal forms.

YU Patent 44,495 describes a process of synthesizing C-S-H to obtain a product which consists of the mineral tobermorite. This product has good mechanical and thermal  
30 insulation properties but includes asbestos and linen fibres.

South African Patent 94/5548 describes a process of  
35 synthesizing C-S-H which contains the mineral xonotlite C<sub>6</sub>S<sub>6</sub>H. Due to the selection of raw materials, the ratios thereof and proper reaction conditions, the formed

xonotlite provides finished products with higher rigidity and lower linear shrinkage at high temperatures and consequently higher thermal stability. The finished products are also free from asbestos fibres which are not  
5 desired and even forbidden lately because of harmful effects on human health.

A xonotlite molecule contains only one crystal water molecule and this differs, for example, from 11YA  
10 tobermorite ( $C_5S_6H_5$ ) which contains five molecules of crystal bound water per molecule. When exposed to high temperatures the phase transformation of C-S-H compound containing more crystal water molecules starts at a considerably lower temperature. For this reason the  
15 presence of the mineral xonotlite decreases linear deformation and increases mechanical properties of the product due to high temperatures. Besides, the addition of a fibrous component and a surfactant affects the formation of a microstructure which increases the  
20 viscosity of the porous material and makes the product resistant to sudden temperature changes.

It is accordingly an object of the present invention to provide an alternative process of producing a C-S-H  
25 product, preferably a C-S-H product which includes the mineral xonotlite  $C_6S_6H$ .

According to the present invention there is provided a process for producing a thermal insulating calcium  
30 hydrosilicate compound comprising:

- providing a source of calcium ions;
- providing a silica compound;
- providing a stabilizing reagent;
- providing a fibrous compound;
- 35 - forming an aqueous mixture of the source of calcium ions the silica compound, the stabilizing reagent and the fibrous component; and

- hydrothermally treating the aqueous mixture at a pressure above 12,5 bars but not over 20 bars in the presence of steam at a temperature from about 190 to 212°C to produce a calcium hydrosilicate compound.

5

The source of calcium ions may comprise lime. In one embodiment of the invention the lime may comprise slaked lime preferably with a CaO content of not less than 72% by weight. Alternatively it may comprise quicklime preferably containing not less than 95% CaO.

10

The silica compound may comprise a compound selected from the group consisting of amorphous silica, silica sand, quartz, quartzite, diatomite and mixtures thereof. In one embodiment it may comprise quartz and the quartz may be quartz which was previously treated at a temperature of not higher than 1000°C and preferably between 800 and 1000°C.

15

- 20 Preferably the silica compound contains not less than 98% (by weight) SiO<sub>2</sub>.

Preferably the silica compound has a particle size of below 63µm and preferably at least 90% of the silica compound has a particle size of below 45µm.

25

The silica compound and source of calcium ions may have a mole ratio of Ca<sup>2+</sup>/SiO<sub>2</sub> of between 0,91 and 1.2.

- 30 Preferably the amount of water in the aqueous mixture is 3 to 4 times higher by weight than the total dry substance.

- The fibrous compound may comprise alkali stable fibres.
- 35 The fibres may be organic, inorganic, natural, synthetic or mixtures of such fibres. Examples of fibres are mineral fibres, alkaline resistant glass fibres,

cellulose fibres, polypropylene fibres, polyethylene fibres, polyester fibres, polyamide fibres, polyacrylonitrile fibres etc.

- 5 Preferably the fibres comprise sulphate fir white cellulose. Alternatively or additionally it comprises alkali resistant glass fibres which preferably have a 10% by weight  $ZrO_2$  content and 5 - 8% by weight  $TiO_2$  content and preferably with a thickness of approximately  $0,025\mu m$   
10 and a length of 15 to  $25\mu m$ .

- The fibrous component may be introduced in an amount of 2 to 8% by weight of the total dry substance. Preferably it is introduced in an amount of 5 to 7% by weight of the  
15 total dry substance.

- The stabilizing reagent is used to stabilize the aqueous mixture and may comprise a surfactant such as alkylphenolpolyglycol ether or a cellulose derivative.  
20 Preferably it comprises a compound selected from the group consisting of carboxymethylcellulose (including salts thereof), metasilicilic acid, active starch, gelatine, alkylhydroxyalkyl cellulose and glass fibres. The carboxymethylcellulose may comprise  
25 Na-carboxymethylcellulose and the alkylhydroxyalkyl-cellulose may comprise ethylhydroxyethylcellulose. Preferably the stabilizing reagent comprises metasilicilic acid or Na-carboxymethylcellulose.

- 30 Preferably the stabilizing reagent is introduced in an amount of 0,2 to 1%, preferably 0,4 to 0,6% by weight of the total dry substance.

- 35 Preferably the lime, silica and stabilizing reagent are mixed with water to form an aqueous mixture; the fibrous compound is also mixed with water to form a separate aqueous mixture; and the two aqueous mixtures are then

mixed together. This mixture may then be introduced into moulds which are then subjected to hydrothermal treatment at a pressure above 12,5 bars but not above 15 bars.

- 5 The hydrothermal treatment may be carried out in an autoclave.

Preferably the hydrothermal treatment is carried out for a period of 10 to 25 hours.

10

The hydrothermally treated product may then be dried, preferably at a temperature of between 60 to 120°C.

- 15 In a preferred embodiment of the invention the process includes the following steps:

- preparation of an aqueous mixture of lime, silica and a stabilizing reagent;
- preparation of a fibrous compound aqueous mixture;
- mixing together both mixtures of the previous steps;
- 20 - pouring of the mixture thus obtained into one or more moulds;
- transfer the one or more moulds containing the mixture into an autoclave and treating it with saturated water steam at a pressure of above 12,5 bars but below 20
- 25 bars to allow the slurry to solidify and crystals to form;
- relieving the pressure in the autoclave to atmospheric pressure and removal of the one or more moulds from the autoclave; and
- 30 - release of the formed product from the one or more moulds and drying the formed product in a drier.

Preferably the calcium hydrosilicate compound includes the mineral xonotlite.

35

According to another aspect of the invention there is provided a product produced by the process substantially



as described hereinabove. The product may comprise calcium hydrosilicate and preferably it includes xonotlite.

- 5 The invention will now be further described with reference to the accompanying non-limiting examples:

Example 1

- 10 The following components were used in the amounts indicated to produce a C-S-H compound containing xonotlite.

	1. Slaked lime	661kg
	2. Quartz	640kg
15	3. Metasilicilic acid	8kg
	4. Sulphate fir white cellulose	90kg
	5. Water	4283l

- 20 The slaked lime with a CaO content of 72% by weight was suspended in 2000 l of water and stirred for 45 minutes. To this was added 513 l of water with 8kg metasilicilic acid and 640kg of quartz. The metasilicilic acid is a stabilizing reagent which stabilizes the mixtures of the components.

- 25 The quartz comprised pulverized quartz sand, containing 98% SiO<sub>2</sub> and the particle size was below 63µm with at least 90% of the quartz having particle size of below 45µm.

- 30 The sulphate fir white cellulose was suspended in 1770 l of water.

- 35 The quartz, metasilicilic acid and lime suspension was added to the aqueous suspension of the cellulose. The resulting slurry was stirred for 60 minutes, and then poured in moulds of dimensions 3,0mx1,2mx0,3m.

- The moulds were then transferred to an autoclave and was hydrothermally treated by the introduction of saturated water steam. The treatment was carried out at a temperature of 198°C. A pressure of 14,9 bars was obtained in the autoclave within 50 minutes and maintained for 14 hours. The autoclave was cooled in order that the pressure was reduced to 7 bars within 3 hours. Within a further 4 hours the pressure was reduced to atmospheric pressure.
- The moulds were removed from the autoclave, the blocks were released and then dried at a temperature of not higher than 120°C to have a moisture content of 10-20%. The product obtained had a bulk density of 321 to 355kg/m<sup>3</sup> a bending strength to 2,7 MPa, with the mineral xonotlite in its structure.

#### Example 2

The following components were used:

- |                                 |       |
|---------------------------------|-------|
| 1. Slaked lime                  | 650kg |
| 2. Quartz                       | 495kg |
| 3. Metasilicilic acid           | 6kg   |
| 4. Sulphate fir white cellulose | 50kg  |
| 5. Glass fibres                 | 20kg  |
| 6. Water                        | 4300l |

Components 1 to 4 were the same as the components described in example 1. Glass fibres were alkali resistant, contained 10% (by weight) ZrO<sub>2</sub> and 5-8%TiO<sub>2</sub> (by weight) and had a thickness of about 0.025µm and a length of about 15-25µm.

The same procedure as set out in example 1 was followed but in this case a pressure of 12.6 bars was maintained for 25 hours. Temperature of saturated water steam was 190°C. The obtained product had a bulk density of 354kg/m<sup>3</sup>, a bending

strength to 2,5MPa and mineral xonotlite prevailed in the structure.

### Example 3

5

The following components were used:

	1. Quicklime	562kg
	2. Quartz	570kg
10	3. Metasilicilic acid	7,9kg
	4. Sulphate fir white cellulose	80kg
	5. Water	4283l

15 The quicklime contained 96% CaO. The quartz and the rest of the components were the same as the components described in example 1.

20 The same procedure as set out in example 1 was carried out, but in this case the hydrothermal treatment was carried out at a temperature of 195°C.

25 A pressure of 14 bars was obtained within 90 minutes and maintained for 14 hours. After that the autoclave was cooled in order that the pressure was reduced to 8 bars within one hour. Within a further 4 hours the pressure was reduced to atmospheric pressure.

30 After drying, the product obtained had the following properties: a bulk density of 335 to 373kg/m<sup>3</sup> a bending strength of 2,5 mPa. The product mainly consisted of cross-linked crystals of xonotlite.

### Example 4

35 The following components were used:

	1. Quicklime	585kg
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10

2. Amorphous silica	580kg
3. Na-carboxymethylcellulose	5kg
4. Sulphate fir white cellulose	85kg
5. Water	4660l

5

The quicklime contained 97% CaO.

Ultra fine amorphous silica was used and it had a silica content and particle size as set out in example 1.

10

The stabilizing reagent was Na-carboxymethylcellulose.

The same procedure as set out in example 1 was carried out, but in this case the hydrothermal treatment was carried out at a temperature of 212°C. A pressure of 19.9 bars was obtained within 90 minutes and maintained for 10 hours. Thereafter the autoclave was cooled for 90 minutes until the pressure was reduced to 8 bars. Within a further 4 hours the pressure was reduced to atmospheric pressure.

20

After drying, the product obtained had the following properties: a bulk density of 320-340kg/m<sup>3</sup>, a bending strength of 3MPa. The product mainly consisted of cross-linked crystals of xonotlite.

25

It will be appreciated that many variations in detail are possible without thereby departing from the scope and spirit of the invention.

CLAIMS

1. A process for producing a thermal insulating calcium hydrosilicate compound comprising:
  - 5 - providing a source of calcium ions;
  - providing a silica compound;
  - providing a stabilizing reagent;
  - providing a fibrous compound;
  - forming an aqueous mixture of the source of calcium  
10 ions the silica compound, the stabilizing reagent and the fibrous component; and
  - hydrothermally treating the aqueous mixture at a pressure above 12,5 bars but not over 20 bars in the presence of saturated steam at a temperature from  
15 about 190 to 212°C to produce a calcium hydrosilicate compound.
2. The process of claim 1 wherein the source of calcium ions comprises lime.  
20
3. The process of claim 2 wherein the lime comprises slaked lime with a CaO content of not less than 72% by weight.
- 25 4. The process of claim 2 wherein the lime comprises quicklime containing not less than 95% CaO.
5. The process of claim 1 wherein the silica compound comprises a compound selected from the group  
30 consisting of amorphous silica, silica sand, quartz, quartzite and diatomite.
6. The process of claim 5 wherein the silica compound comprises quartz previously treated at a temperature  
35 of not higher than 1000°C.

7. The process of claim 1 wherein the silica compound contains not less than 98% (by weight)  $\text{SiO}_2$ .
- 5 8. The process of claim 1 wherein the silica compound has a particle size of below  $63\mu\text{m}$  and at least 90% of the silica compound has a particle size of below  $45\mu\text{m}$ .
- 10 9. The process of claim 1 wherein the silica compound and source of calcium ions have a mole ratio of  $\text{Ca}^{2+}/\text{SiO}_2$  of between 0,91 and 1.2.
- 15 10. The process of claim 1 wherein the amount of water in the aqueous mixture is 3 to 4 times higher by weight than the total dry substance.
- 20 11. The process of claim 1 wherein the fibrous compound comprises sulphate fir white cellulose and/or inorganic fibres.
12. The process of claim 11 wherein the fibrous compound is introduced in an amount of 2 to 8% by weight of the total dry substance.
- 25 13. The process of claim 12 wherein the fibrous compound is introduced in an amount of 5 to 7% by weight of the total dry substance.
- 30 14. The process of claim 1 wherein the stabilizing reagent comprises a compound selected from the group consisting of carboxymethylcellulose (including salts thereof), metasilicilic acid, active starch, gelatine, ethylhydroxyethyl-cellulose and glass fibre.
- 35 15. The process of claim 14 wherein the stabilizing reagent comprises metasilicilic acid.

16. The process of claim 14 wherein the stabilizing reagent comprises Na-carboxymethylcellulose.
- 5 17. The process of any one of the preceding claims wherein the stabilizing reagent is introduced in an amount of 0,2 to 1% by weight of the total dry substance.
- 10 18. The process of claim 2 wherein the lime, silica and stabilizing reagent are mixed with water to form an aqueous mixture; the fibrous compound is also mixed with water to form a separate aqueous mixture; and the two aqueous mixtures are then mixed together.
- 15 19. The process of claim 1 wherein the hydrothermal treatment is carried out in an autoclave.
- 20 20. The process of claim 19 wherein the hydrothermal treatment is carried out for a period of 10 to 25 hours.
21. The process of any one of the preceding claims wherein the hydrothermally treated product is dried at a temperature of between 60 to 120°C.
- 25 22. A process for producing a thermal insulating calcium hydrosilicate compound comprising:
- preparation of an aqueous mixture of lime, silica and a stabilizing reagent;
  - preparation of a fibrous compound aqueous mixture;

30 - mixing together both mixtures of the previous steps;

  - pouring of the mixture thus obtained into one or more moulds;
  - transfer the one or more moulds containing the mixture into an autoclave and treating it with water

35 steam at a pressure of above 12,5 bars but below 20 bars to allow the slurry to solidify and crystals to form;

- relieving the pressure in the autoclave to atmospheric pressure and removal of the one or more moulds from the autoclave; and
- release of the formed product from the one or more moulds and drying the formed product.

5

23. The process of any one of the preceding claims wherein the calcium hydrosilicate compound includes the mineral xonotlite.

10

24. A product produced by any one of the preceding claims.

25. Xonotlite produced by any one of claims 1 to 23.



## INTERNATIONAL SEARCH REPORT

national Application No

PCT/GB 96/01033

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 6 C04B40/02 C04B28/20

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C04B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE,A,36 41 823 (CSP CHEMIE ENTWICKLUNGSGESELLS) 16 June 1988	1-5,7, 9-14,18, 19,22-25
A	see column 1, line 45 - column 3, line 17; claims; figure 1; example see the whole document	6,8, 15-17, 20,21
Y	FR,A,2 278 647 (LILLE INST CATHOLIQUE ARTS M) 13 February 1976 see the whole document	1-5,11, 14,19, 22-25

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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Fax: (+ 31-70) 340-3016

Authorized officer

Harbron, J

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	DATABASE WPI Section Ch, Week 9528 Derwent Publications Ltd., London, GB; Class L02, AN 95-212815 XP002011440 & JP,A,07 126 083 (KURARAY CO LTD) , 16 May 1995 see abstract	1-5,11, 14,19, 22-25
A	--- GB,A,1 269 960 (OWENS-CORNING) 12 April 1972	
A	--- NL,A,8 100 680 (WARRIOR INSULATION) 1 September 1982 -----	

**INTERNATIONAL SEARCH REPORT**

Information on patent family members

national Application No

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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
DE-A-3641823	16-06-88	DE-A- 3783444 EP-A- 0270797	18-02-93 15-06-88
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GB-A-1269960	12-04-72	NONE	
NL-A-8100680	01-09-82	NONE	